INTERNATIONAL SEARCH REPORT

International Application No
PCT/EP2004/012334

A. CLASSIFICATION OF SUBJECT MATTER
IPC 7 G01M3/20

According to International Patent Classification (IPC) or to both national classification and IPC

B. FIELDS SEARCHED

Minimum documentation searched (classification system followed by classification symbols)
IPC 7 G01M

Documentation searched other than minimum documentation to the extent that such documents are included in the fields searched

Electronic data base consulted during the international search (name of data base and, where practical, search terms used)

EPO-Internal, WPI Data, PAJ

C. DOCUMENTS CONSIDERED TO BE RELEVANT

Category °	Citation of document, with indication, where appropriate, of the relevant passages	Relevant to claim No.
A	DE 101 22 733 A1 (INFICON GMBH) 14 November 2002 (2002-11-14) paragraph '0005!; claim 1; figure 1 -----	1
A	WO 02/47189 A (SIEMENS AKTIENGESELLSCHAFT; MATTEJAT, ARNO; VOITLEIN, OTTMAR) 13 June 2002 (2002-06-13) abstract; figure 1 -----	1
A	EP 0 395 965 A (LUZ INDUSTRIES ISRAEL LTD) 7 November 1990 (1990-11-07) abstract; figure 1 -----	1
A	EP 0 503 925 A (AGENCY OF INDUSTRIAL SCIENCE & TECHNOLOGY MINISTRY OF INTERNATIONAL TR) 16 September 1992 (1992-09-16) column 4, line 56 - column 5, line 45; figure 3 -----	1

☐ Further documents are listed in the continuation of box C.

☒ Patent family members are listed in annex.

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Date of the actual completion of the international search	Date of mailing of the international search report
11 April 2005	19/04/2005

Name and mailing address of the ISA	Authorized officer
European Patent Office, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Trique, M

INTERNATIONAL SEARCH REPORT

Information on patent family members

International Application No
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Patent document cited in search report	Publication date	Patent family member(s)	Publication date
DE 10122733 A1	14-11-2002	WO 02090917 A2	14-11-2002
WO 0247189 A	13-06-2002	DE 10060626 A1	20-06-2002
		CA 2437004 A1	13-06-2002
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		DE 50105151 D1	24-02-2005
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		JP 2004515889 T	27-05-2004
		US 2005003245 A1	06-01-2005
EP 0395965 A	07-11-1990	US 4892142 A	09-01-1990
		AU 5451090 A	08-11-1990
		BR 9002104 A	13-08-1991
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		JP 6258170 A	16-09-1994
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		DE 69200871 D1	26-01-1995
		DE 69200871 T2	27-07-1995
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		KR 137128 B1	29-04-1998
		US 5261268 A	16-11-1993

INTERNATIONALER RECHERCHENBERICHT

Internationales Aktenzeichen
PCT/EP2004/012334

A. KLASSIFIZIERUNG DES ANMELDUNGSGEGENSTANDES
IPK 7 G01M3/20

Nach der Internationalen Patentklassifikation (IPK) oder nach der nationalen Klassifikation und der IPK

B. RECHERCHIERTE GEBIETE

Recherchierter Mindestprüfstoff (Klassifikationssystem und Klassifikationssymbole)
IPK 7 G01M

Recherchierte aber nicht zum Mindestprüfstoff gehörende Veröffentlichungen, soweit diese unter die recherchierten Gebiete fallen

Während der internationalen Recherche konsultierte elektronische Datenbank (Name der Datenbank und evtl. verwendete Suchbegriffe)

EPO-Internal, WPI Data, PAJ

C. ALS WESENTLICH ANGESEHENE UNTERLAGEN

Kategorie°	Bezeichnung der Veröffentlichung, soweit erforderlich unter Angabe der in Betracht kommenden Teile	Betr. Anspruch Nr.
A	DE 101 22 733 A1 (INFICON GMBH) 14. November 2002 (2002-11-14) Absatz '0005!; Anspruch 1; Abbildung 1 -----	1
A	WO 02/47189 A (SIEMENS AKTIENGESELLSCHAFT; MATTEJAT, ARNO; VOITLEIN, OTTMAR) 13. Juni 2002 (2002-06-13) Zusammenfassung; Abbildung 1 -----	1
A	EP 0 395 965 A (LUZ INDUSTRIES ISRAEL LTD) 7. November 1990 (1990-11-07) Zusammenfassung; Abbildung 1 -----	1
A	EP 0 503 925 A (AGENCY OF INDUSTRIAL SCIENCE & TECHNOLOGY MINISTRY OF INTERNATIONAL TR) 16. September 1992 (1992-09-16) Spalte 4, Zeile 56 - Spalte 5, Zeile 45; Abbildung 3 -----	1

☐ Weitere Veröffentlichungen sind der Fortsetzung von Feld C zu entnehmen

☒ Siehe Anhang Patentfamilie

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Datum des Abschlusses der internationalen Recherche	Absendedatum des internationalen Recherchenberichts
11. April 2005	19/04/2005
Name und Postanschrift der Internationalen Recherchenbehörde Europäisches Patentamt, P.B. 5818 Patentlaan 2 NL – 2280 HV Rijswijk Tel. (+31-70) 340-2040, Tx. 31 651 epo nl, Fax: (+31-70) 340-3016	Bevollmächtigter Bediensteter Trique, M

INTERNATIONALER RECHERCHENBERICHT

Angaben zu Veröffentlichungen, die zur selben Patentfamilie gehören

Internationales Aktenzeichen
PCT/EP2004/012334

Im Recherchenbericht angeführtes Patentdokument	Datum der Veröffentlichung	Mitglied(er) der Patentfamilie	Datum der Veröffentlichung
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WO 0247189 A	13-06-2002	DE 10060626 A1	20-06-2002
		CA 2437004 A1	13-06-2002
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EP 0395965 A	07-11-1990	US 4892142 A	09-01-1990
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		US 5261268 A	16-11-1993

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Method for operating a hydrogen test leak unit

The invention relates to a method for operating a hydrogen test leak unit comprising a chamber defined by a membrane, into which chamber a test gas mixture of hydrogen and at least one added gas is introduced.

Test leak units are used for calibrating leakage flow meters. A test leak unit comprises a container in which a volume of a test gas or a test gas mixture is kept at a controlled pressure. The chamber includes an outlet connected to a capillary or any other defined test leak leading into the surrounding. For pressure control purposes, the chamber comprises a membrane which is supported by a spring, and which, upon deflection, actuates a control valve which switches on and off a gas-supplying pressure source.

Hydrogen may not be used as a pure gas in test leak units due to its explosiveness. Therefore, normally an oxygen-free hydrogen/nitrogen mixture is used as a test gas mixture (forming gas). However, the membrane sealing the chamber of the pressure control valve is not gas-impermeable. It rather offers different permeabilityies to different gases. Consequently, the hydrogen escapes more quickly from the chamber than nitrogen such that the nitrogen concentration in the chamber increases. Normally, the test gas mixture comprises 95 % N_2 and 5 % H_2. The hydrogen content can amount to up to 10 %. This leads to an explosion risk.

It is an object of the invention to provide a method for operating a hydrogen test leak unit which offers a high time constancy of the leak rate.

The method according to the invention is defined in claim 1. According to the invention, a gas is selected as an "added gas" which has a permeation coefficient relative to the material of the membrane ranging between 50 % and 200 % of that of hydrogen.

The invention is based on the idea that the components of the test gas mixture should have approximately the same permeation coefficients relative to the membrane material in order to prevent demixing due to permeation. The permeation coefficients depend to a large extent, but not exclusively, on the molecule size. The hydrogen molecule is relatively small and generally has relatively high permeation coefficients. This applies, mutatis mutandis, to helium. On the other hand, it has been found that even relatively large molecules, such as CO_2, CH_4 or C_2H_6, may offer a high permeability.

Helium has turned out to be a particularly suitable gas to be added to hydrogen. Experiments have shown that permeation through a membrane proceeds at essentially the same rate for hydrogen and helium. This combination offers the advantage that the hydrogen content can be reduced to approximately 5 %. This is of importance with regard to hazardous substance categories. Another boundary condition requires that the useful life of test leak units should be one year. So far, realization of a hydrogen content of 5 % has not been feasible since too much gas escaped through the required bypass capillary, and it has not been possible to comply with the useful life.

Suitable gases to be added are helium (He), carbon dioxide (CO_2), methane (CH_4) and ethane (C_2H_6). This list is however not complete.

The gas to be added should be oxygen-free to prevent it from forming, together with hydrogen, an explosive mixture. This is however not a necessary condition. By reducing the hydrogen content, the explosiveness of the test gas mixture can be decreased.

An embodiments of the invention will now be described in greater detail with reference to the drawings in which:

Fig. 1 shows a functional diagram of the hydrogen test leak unit; and

Fig. 2 shows a schematic configuration of the hydrogen test leak unit.

As shown in Fig. 1, a test gas mixture 10 is introduced into a pressure control volume 11. The pressure control volume 11 comprises a pressure reducing valve provided with a control membrane 12. A pressure control valve 13 adjusts, with the aid of the control membrane 12, the pressure in the pressure control volume 11 to a constant value. The pressure control valve is connected with a leak rate-determining capillary 14 from which the leakage flow 15 escapes at the test gas leak rate.

The pressure control volume 11 or the pressure control valve 13 is connected with a bypass capillary 16 which leads a bypass flow 17 into the surrounding for the purpose of ensuring a permanent flow through the pressure control volume 11 to prevent demixing.

Fig. 2 shows the pressure controller 20 to which the test gas mixture 10 is supplied. The pressure controller 20 comprises a chamber 21 into which the test gas mixture is introduced. This chamber is sealed with a membrane 22. The membrane 22 is made from an elastomeric material. It is supported by a spring 23 which counteracts the pressure prevailing in the chamber 21. The pressure control valve 24 comprises a duct 25 for delivering the highly pressurized test gas mixture into the chamber 21. The outlet of the duct 25 is opened and closed by a movable valve element 26. Via a rod 27 defining a two-arm lever, the valve element 26 is connected with a coupling means 28 which connects one of the lever arms with the membrane 22. The deflection movements of the membrane 22 cause the pressure control valve 24 to be opened or closed. In this manner, a pressure corresponding to the force of the spring 23 is adjusted and maintained in the chamber 21.

Fig. 2 further shows the test leak outlet 30 which is connected with a leak rate-determining capillary (not shown) leading to the surrounding.

Further, a bypass capillary 31 is provided at the chamber 21, which bypass capillary permanently leads a leak flow into the surrounding.

The space below the membrane 22 is defined by a container 33 which supports the spring 23 and comprises a vent opening 34.

The small dots in Fig 2 represent the mixed-gas volume and the bigger dots represent the hydrogen molecules. The hydrogen molecules diffuse through the membrane 22 out of the chamber 21 and travel through the vent opening 34 into the surrounding.

The membrane 22 is a polymeric membrane. In one embodiment it is made from polybutadiene-co-acrylonitrile (Perbunan 18). In another embodiment it is made from 73/27 Perbunan.

The following table shows the permeation coefficient P for the stated membrane materials. In the table given below (from Yasuda, H., Stannet, V.: Polymer Handbook, J. Wiley & Sons, New York 1975) the permeation coefficient P has the following dimension

$$[cm^3\ (STP)\ cm\ cm^{-2}\ s^{-1}\ (cm\ Hg)^{-1}]$$

Table

Membrane material		
Poly(butadiene-co-acrylonitrile) 80/20		$P \times 10^{10}$
	He	16.9
	H_2	25.2
	N_2	2.52
	O_2	8.16
	CO_2	63.1
73/27 (Perbunan)		
	He	12.2

H_2	15.9
N_2	1.06
O_2	3.85
CO_2	30.8
C_2H_2	24.9
C_3H_8	77.7

Depending on the membrane material, the permeation coefficient P for hydrogen is 25.2 or 15.9. The gas to be added is selected such that its permeation coefficient ranges between 50 and 200 % of that of hydrogen. For this purpose, other gases which are not indicated in the table can also be used as added gases. Preferably, the stated range lies between 50 % and 150 %.

CLAIMS

1. A method for operating a hydrogen test leak unit comprising a chamber (21) defined by a membrane (22), into which chamber a test gas mixture of hydrogen and at least one added gas is introduced,

   c h a r a c t e r i z e d   i n   t h a t

   the added gas has a permeation coefficient (P) relative to the material of the membrane (22) ranging between 50 % and 200 % of that of hydrogen.

2. The method according to claim 1, characterized in that helium, carbon dioxide or methane is selected as added gas.

3. The method according to claim 1 or 2, characterized in that the added gas is oxygen-free.

4. The method according to one of claims 1-3, characterized in that the hydrogen content of the test gas mixture is lower than 10 %.

ABSTRACT OF THE DISCLOSURE

Method for operating a hydrogen test leak unit

For operating a hydrogen test leak unit comprising a chamber (21) defined by a membrane (22), a test gas mixture of hydrogen and an added gas is used. The added gas has a permeation coefficient relative to the material of the membrane (22) ranging between 50 % and 200 % of that of hydrogen. In this manner, demixing of the test gas mixture is limited and significant concentration variations in the chamber (21) are prevented. Preferably, helium is used as added gas.

(Fig. 2)

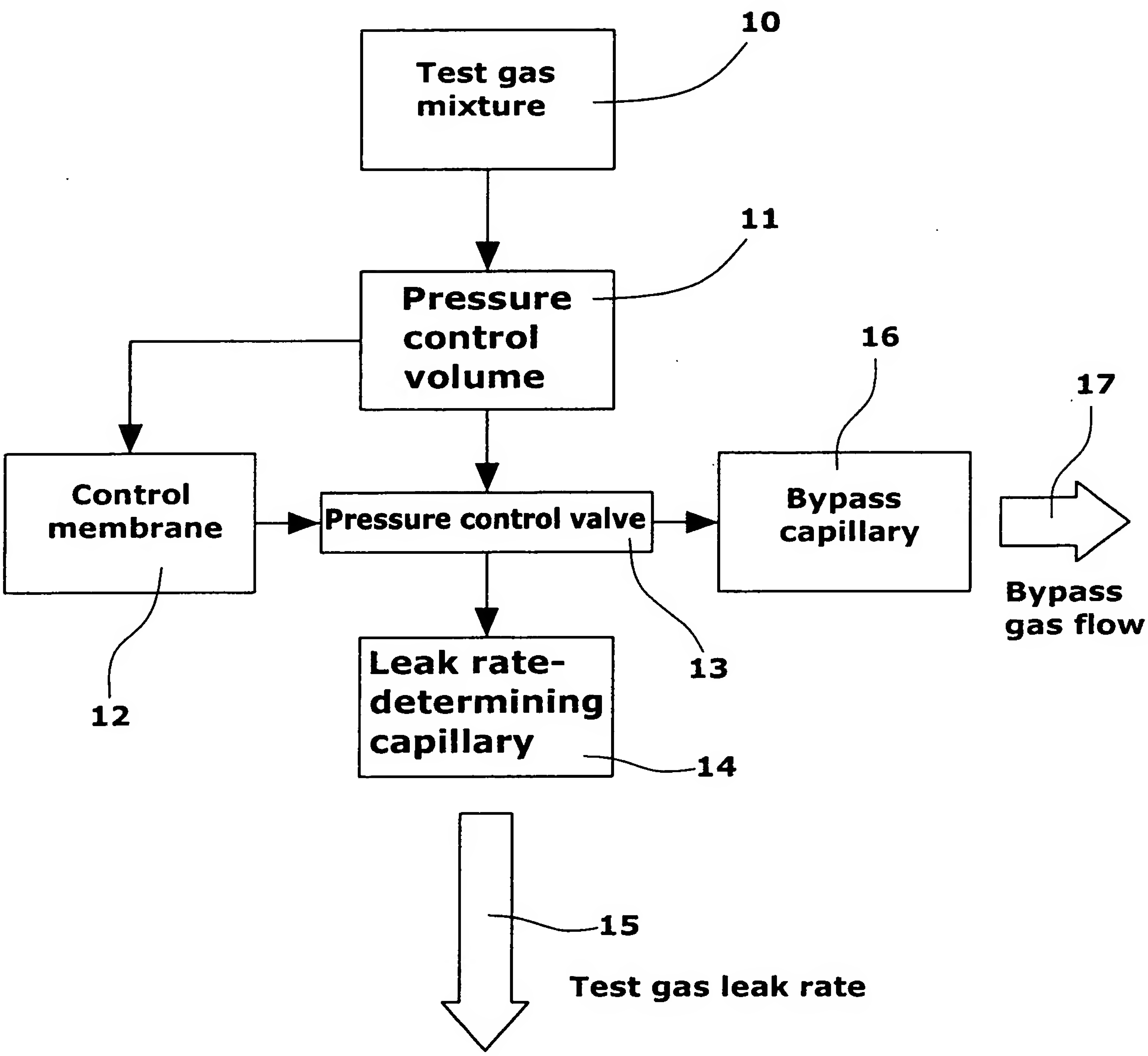
10
Test gas mixture
11
Pressure control volume
16
17
Control membrane
Pressure control valve
Bypass capillary
Bypass gas flow
12
13
Leak rate-determining capillary
14
15
Test gas leak rate

Fig.1

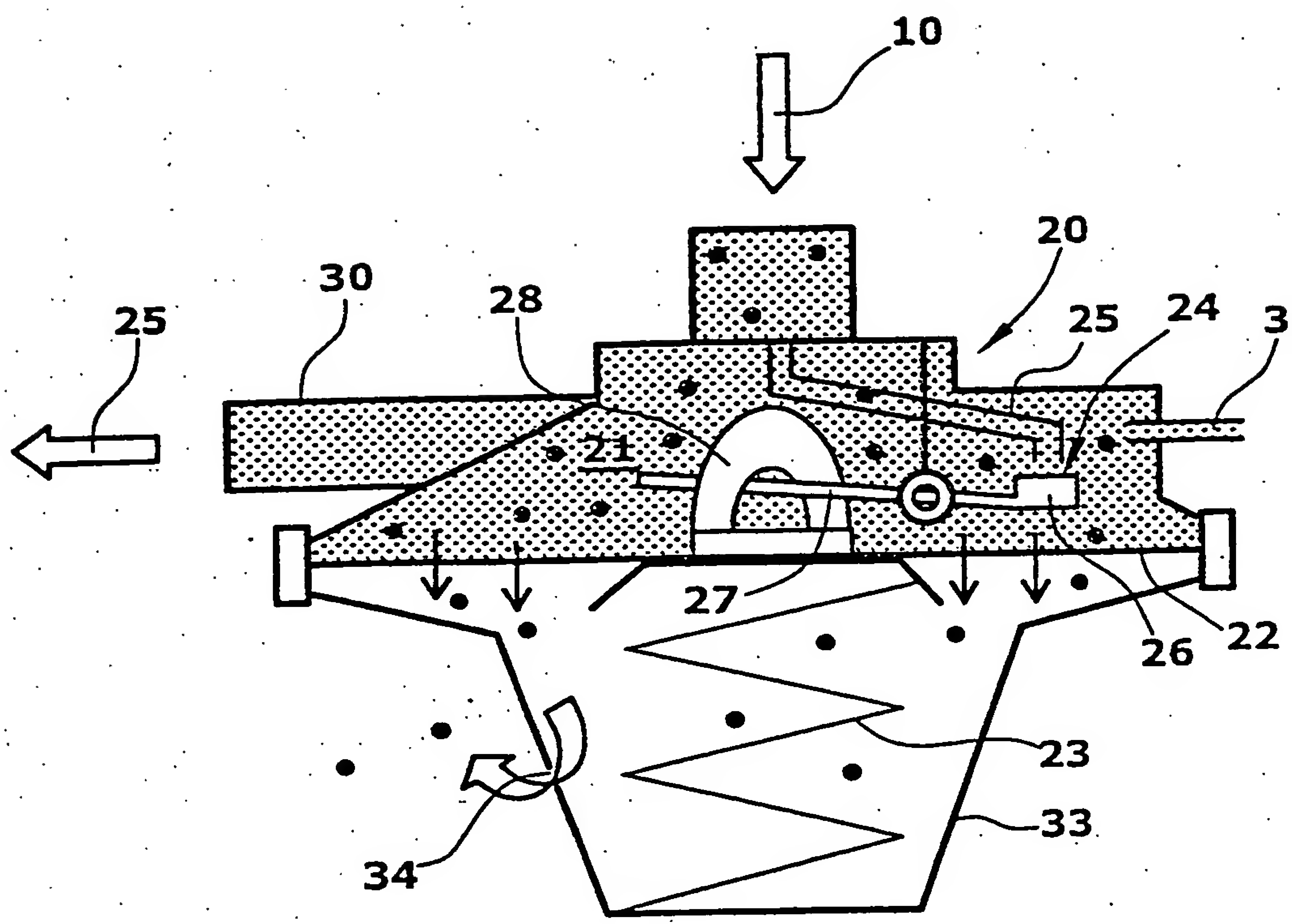

Fig.2